

**PCT**WORLD INTELLECTUAL PROPERTY ORGANIZATION  
International Bureau

## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<b>(51) International Patent Classification 6 :</b> <b>A01N 65/00</b>	<b>A1</b>	<b>(11) International Publication Number:</b> <b>WO 99/53763</b> <b>(43) International Publication Date:</b> 28 October 1999 (28.10.99)
<b>(21) International Application Number:</b> PCT/IN99/00011 <b>(22) International Filing Date:</b> 31 March 1999 (31.03.99) <b>(30) Priority Data:</b> 827/Mas/98 17 April 1998 (17.04.98) IN <b>(71)(72) Applicants and Inventors:</b> SUBBA RAO, Pillarisetti, Venkata [IN/IN]; 41/3, 13th Cross, Malleswaram, Bangalore 560 003 (IN). ANNADURAI, Ramasamy, Sambasivam [IN/IN]; 1840, 10th Main Road, 34th Cross, Banshankari II Stage, Bangalore 560 070 (IN). SRINIVAS, Malladi [IN/IN]; 319, IC Cross, II Phase, 6th Block, Banshankari III Stage, Bangalore 560 085 (IN). <b>(74) Agents:</b> ANAND, Pravin et al.; Anand & Anand Advocates, B-41, Nizammuddin East, New Delhi 110 013 (IN).		<b>(81) Designated States:</b> AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report.</i>
<b>(54) Title:</b> AN ENVIRONMENT FRIENDLY ACARICIDE FORMULATION  <b>(57) Abstract</b>  This invention relates to an environment friendly acaricide formulation composition for the control of house dust mites comprising: plant derived acaricidal agent 0.01–0.1 % wt./vol.; plant derived disinfectant agent 0.3–3 % wt./vol.; plant derived protein denaturant 0.1–2 % wt./vol.; fungistat agent 0.1–3 % wt./vol.; dispersing agent (alcohol) 99.69–91.9 % wt./vol.		

**FOR THE PURPOSES OF INFORMATION ONLY**

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AL	Albania	ES	Spain	LS	Lesotho	SI	Slovenia
AM	Armenia	FI	Finland	LT	Lithuania	SK	Slovakia
AT	Austria	FR	France	LU	Luxembourg	SN	Senegal
AU	Australia	GA	Gabon	LV	Latvia	SZ	Swaziland
AZ	Azerbaijan	GB	United Kingdom	MC	Monaco	TD	Chad
BA	Bosnia and Herzegovina	GE	Georgia	MD	Republic of Moldova	TG	Togo
BB	Barbados	GH	Ghana	MG	Madagascar	TJ	Tajikistan
BE	Belgium	GN	Guinea	MK	The former Yugoslav Republic of Macedonia	TM	Turkmenistan
BF	Burkina Faso	GR	Greece	ML	Mali	TR	Turkey
BG	Bulgaria	HU	Hungary	MN	Mongolia	TT	Trinidad and Tobago
BJ	Benin	IE	Ireland	MR	Mauritania	UA	Ukraine
BR	Brazil	IL	Israel	MW	Malawi	UG	Uganda
BY	Belarus	IS	Iceland	MX	Mexico	US	United States of America
CA	Canada	IT	Italy	NE	Niger	UZ	Uzbekistan
CF	Central African Republic	JP	Japan	NL	Netherlands	VN	Viet Nam
CG	Congo	KE	Kenya	NO	Norway	YU	Yugoslavia
CH	Switzerland	KG	Kyrgyzstan	NZ	New Zealand	ZW	Zimbabwe
CI	Côte d'Ivoire	KP	Democratic People's Republic of Korea	PL	Poland		
CM	Cameroon	KR	Republic of Korea	PT	Portugal		
CN	China	KZ	Kazakhstan	RO	Romania		
CU	Cuba	LC	Saint Lucia	RU	Russian Federation		
CZ	Czech Republic	LI	Liechtenstein	SD	Sudan		
DE	Germany	LK	Sri Lanka	SE	Sweden		
DK	Denmark	LR	Liberia	SG	Singapore		
EE	Estonia						

### *An Environment Friendly Acaricide Formulation*

This invention relates to an environment friendly acaricide formulation for the control of house dust mite population in the domestic environment responsible for high incidence of respiratory allergies and process of preparing the same.

#### 5 BACKGROUND

The major culprits for dust allergy are mites (*Dermatophagoides farinae* and *D. pteronyssinus*) prevalent in house dust. Universally, dust mites are minute co-inhabitants in almost every household and can not be seen with the naked eye. They are found in almost all home furnishing textiles and their favourite places are  
10 mattresses, cushions, carpets, upholstery and soft toys. The reactions in hypersensitive people range from itchy and watery eyes, repeated sneezes and running nose, cough and bronchial asthma to childhood eczema. The dust on which they thrive may comprise cotton, wool lint, animal and human dander, crumbs, pollens, molds, etc.

15 House dust mites principally feed on human scales which are primarily found in mattresses, Bedding, carpets etc. During occupation, the temperature and humidity of the human body provides an ideal microclimate in the mattresses for dust mites. Development from egg through larva, protonymph, tritonymph to adult requires about a month in cultures, under optimum conditions. An adult mite can  
20 live up to three months. Their food comprises of protein particles and fungi present in the dust.

Mites may occasionally become airborne during bed-making. It has also been demonstrated that they secrete or release some allergens during bed-making. The allergen may comprise mites, eggs, dead mites and their excreta. A gram of dust  
25 mite may contain up to 1000 mites.

Most particles of the faeces, whose physical properties are similar to pollen are deposited on the nasal mucosa and carried to the lungs causing localized inflammatory responses because of the high concentration of allergen.

Control of mite population in the domestic environment is the best method of preventing house dust allergy. The degree of cleanliness determines the number of house dust mites and the allergen level. Common control measures include vacuum cleaning, treating the carpets and bed spreads with insecticides, acaricides and fungicides. Reducing the mite population by interfering with the food chain has also been practised. However, a safe, environment friendly and effective formulation based on natural products for the control of house dust mite is not yet commercially available.

### PRIOR ART

A few formulations are commercially available like Acardust, Acarosan, Allerbiocid etc., containing benzyl benzoate, the chief acaricide agent in these formulation is toxic at higher concentrations to humans as well a pets. As the effective concentration of benzyl benzoate used in these formulations is very high, its wide spread use as a domestic acaricide could be harmful.

In one acaricidal formulation, derivatives of phenols in combination with several natural oils have been used as the active acaricide agent in combination with an antibiotic Natamycin as a fungicide. But the wide spread use of phenolic derivatives and essential oils is not safe from physiological and odour point of view. Moreover, the fungicide as such can not destroy the mites.

Apart from this, a few chemicals like benzyl alcohol, primiphos methyl, dibutyl phthalate, gama-hexachlorocyclohexane and diethyl-m-toluamide have been reported in literature as miticides. But from the toxicology and environmental safety point of view their use is not recommended.

Accordingly, the **object** of this invention is to provide an environment friendly acaricide formulation for domestic use which should have the following characteristics:

- All the chemicals used should be safe from the toxicology point of view.

- Should have multiple modes of action i.e.. it should control the mite population, prevent the growth of fungi, reduce the existing allergen levels, act as a disinfectant as well as prevent the mites from developing resistance to these chemicals.

5                   - Should not have an offensive odour.

Further, re-establishment of house dust mites after treatment with acaricides is the common problem due to the existence of nymph and eggs. Moreover, the miticide cannot reach the deeper layers of carpets and upholstery. Accordingly, the second **object** of this invention is to control the mites and prevent its re-  
10 establishment by preparing the composition which can not only kill the adult mites but also be a ovicide and a larvicide.

To achieve the said objectives this invention provides an environment friendly acaricide formulation for the control of house dust mites comprising :

- |    |   |                                  |   |                     |
|----|---|----------------------------------|---|---------------------|
|    | - | plant derived acaricidal agent   | - | 0.01-0.1 % wt./vol. |
| 15 | - | plant derived disinfectant agent | - | 0.1-3 % wt./vol.    |
|    | - | plant derived protein denaturant | - | 0.1-2 % wt./vol.    |
|    | - | fungistat agent                  | - | 0.1-3% wt./vol.     |
|    | - | dispersing agent (alcohol)       | - | 99.69-91.9% wt./vol |

The plant derived acaricidal agent is neem seed kernel extract containing  
20 azadirachtin / azadirachtin A of 2-90 % enrichment and preferably of 20-35 % enrichment.

The neem seed kernel extract contains limonoids like nimbin, salannin, desacetylnimbin, desacetylsalannin, nimbandiol, azadirachtin-B and salannolacetate for preventing the mites from developing resistance against the  
25 active ingredient.

The plant derived disinfectant agent is an alcoholic extract of resins like stryax benzoin and the plant derived protein denaturant is plant polyphenols like

tannic acid, condensed tannins, phenolic compounds like gallic acid and phloroglucinol.

The fungistat agents are fungicides used in food industry like natamycin, nipagin and the dispersing agents are ethanol, methanol and isopropyl alcohol.

- 5 The ingredients viz. plant derived acaricidal agent, plant derived disinfectant agent, plant derived protein denaturant and fungistat agent of this composition are solids which are dissolved in an alcoholic solvent (dispersing agent) to give a clear pale brown coloured solution.

10 The invention will now be described with reference to the following examples.

#### EXAMPLE – 1

S. No.	Ingredients	Weight/volume (%)
1.	Neem seed kernel extract containing azadirachtin of 20% enrichment	0.1
2.	Alcoholic extract of benzoin resin	3.0
3.	Tannic acid	1.0
4.	Nipagin	1.0
5.	Ethanol	94.9

#### EXAMPLE – 2

15

S. No.	Ingredients	Weight/volume (%)
1.	Neem seed kernel extract containing azadirachtin of 35% enrichment	0.1
2.	Alcoholic extract of benzoin resin	3.0
3.	Tannic acid	1.0
4.	Nipagin	1.0
5.	Ethanol	94.9

**EXAMPLE – 3**

S. No.	Ingredients	Weight/volume (%)
1.	Neem seed kernel extract containing azadirachtin of 90% enrichment	0.1
2.	Alcoholic extract of benzoin resin	3.0
3.	Tannic acid	1.0
4.	Nipagin	1.0
5.	Ethanol	94.9

**EXAMPLE – 4**

5

S. No.	Ingredients	Weight/volume (%)
1.	Neem seed kernel extract containing azadirachtin of 2% enrichment	0.1
2.	Alcoholic extract of benzoin resin	3.0
3.	Tannic acid	1.0
4.	Nipagin	1.0
5.	Ethanol	94.9

**EXAMPLE – 5**

S. No.	Ingredients	Weight/volume (%)
1.	Neem seed kernel extract containing azadirachtin of 20% enrichment	0.1
2.	Alcoholic extract of benzoin resin	3.0
3.	Tannic acid	1.0
4.	Nipagin	1.0
5.	Isopropyl alcohol	94.9

**EXAMPLE – 6**

S. No.	Ingredients	Weight/volume (%)
1.	Neem seed kernel extract containing azadirachtin of 35% enrichment	0.1
2.	Alcoholic extract of benzoin resin	3.0
3.	Tannic acid	1.0
4.	Nipagin	1.0
5.	Isopropyl alcohol	94.9

**EXAMPLE – 7**

5

S. No.	Ingredients	Weight/volume (%)
1.	Neem seed kernel extract containing azadirachtin of 90% enrichment	0.1
2.	Alcoholic extract of benzoin resin	3.0
3.	Tannic acid	1.0
4.	Nipagin	1.0
5.	Isopropyl alcohol	94.9

**EXAMPLE – 8**

S. No.	Ingredients	Weight/volume (%)
1.	Neem seed kernel extract containing azadirachtin of 2% enrichment	0.1
2.	Alcoholic extract of benzoin resin	3.0
3.	Tannic acid	1.0
4.	Nipagin	1.0
5.	Isopropyl alcohol	94.9



**Conclusion:**

Composition in Table 2 is very efficient in terms of speed of action and mite elimination. The total adult population is immobilized within an hour of treatment. A biweekly spray of 200 µl/100mg of the culture is required for 8 weeks to  
5 completely eliminate the population. Reestablishment on treated areas is totally prevented after 8 weeks. After eradication a biweekly prophylactic spray can contain population build up.

**We claim:**

1. An environment friendly acaricide formulation composition characterized by:
 

- plant derived acaricidal agent	- 0.01-0.1 % wt./vol.
- plant derived disinfectant agent	- 0.1-3 % wt./vol.
5    - plant derived protein denaturant	- 0.1-2 % wt./vol.
- fungistat agent	- 0.1-3% wt./vol.
- dispersing agent (alcohol)	- 99.69-91.9% wt./vol
2. Formulation as claimed in claim 1 wherein the plant derived acaricidal agent is neem seed kernel extract containing azadirachtin of 2-90 % enrichment.
- 10    3. Formulation as claimed in claim 1 wherein the plant derived acaricidal agent is neem seed kernel extract containing azadirachtin A of 2-90 % enrichment.
4. Formulation composition as claimed in claim 1 wherein the plant derived acaricidal agent is neem seed kernel extract containing azadirachtin / azadirachtin A preferably of 20-35 % enrichment.
- 15    5. Formulation as claimed in claim 2 wherein the neem seed kernel extract contains limonoids like nimbin, salannin, desacetylnimbin, desacetylsalannin, nimbandiol, azadirachtin-B and salannolacetate for preventing the mites from developing resistance against the active ingredient.
6. Formulation as claimed in claim 1 wherein the plant derived disinfectant agent is an alcoholic extract of resins like stryax benzoin.
- 20    7. Formulation as claimed in claim 1 wherein the plant derived protein denaturant is plant polyphenols like tannic acid, condensed tannins, phenolic compounds like gallic acid and phloroglucinol.
8. Formulation as claimed in claim 1 wherein the fungistat agents are
- 25    fungicides used in food industry like natamycin, nipagin.
9. Formulation as claimed in claim 1 wherein the dispersing agents are ethanol, methanol, isopropyl alcohol.

# INTERNATIONAL SEARCH REPORT

International application No.  
PCT/IN 99/00011

<b>A. CLASSIFICATION OF SUBJECT MATTER</b> IPC <sup>6</sup> : A 01 N 65/00 According to International Patent Classification (IPC) or to both national classification and IPC		
<b>B. FIELDS SEARCHED</b> Minimum documentation searched (classification system followed by classification symbols) IPC <sup>6</sup> : A 01 N Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) WPI		
<b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5 405 612 A (LOCKE et al.), 11 April 1995 (11.04.95), claims.	1-5
A	EP 0 405 291 A1 (W.R.GRACE & CO.), 02 January 1991 (02.01.91), example 5; claims 8,9.	1-5,9
A	DATABASE WPI ON EPOQUE, week 9408, London: Derwent Publications Ltd., AN 94-061960, class B 04m JP 6016515 A (NIPPON KAYAKU KK), abstract.	1-5
A	DE 195 32 447 A1 (REMBOLD), 06 March 1997 (06.03.97), example 1; claims 1-5,15,17,18.	1-5
----		
<input type="checkbox"/> Further documents are listed in the continuation of Box C. <input checked="" type="checkbox"/> See patent family annex.		
* Special categories of cited documents: „A“ document defining the general state of the art which is not considered to be of particular relevance „E“ earlier application or patent but published on or after the international filing date „L“ document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) „O“ document referring to an oral disclosure, use, exhibition or other means „P“ document published prior to the international filing date but later than the priority date claimed „T“ later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention „X“ document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone „Y“ document of particular relevance: the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art „&“ document member of the same patent family		
Date of the actual completion of the international search 28 June 1999 (28.06.99)		Date of mailing of the international search report 03 August 1999 (03.08.99)
Name and mailing address of the ISA/AT Austrian Patent Office Kohlmarkt 8-10; A-1014 Vienna Facsimile No. 1/53424/200		Authorized officer Schnass Telephone No. 1/53424/217

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/IN 99/00011

In Recherchenbericht angeführtes Patentedokument Patent document cited in search report Document de brevet cité dans le rapport de recherche	Datum der Veröffentlichung Publication date Date de publication	Mitglied(er) der Patentfamilie Patent family member(s) Membre(s) de la famille de brevets	Datum der Veröffentlichung Publication date Date de publication
US A 5405612	11-04-1995	AT E 111307 AU A1 62627/90 AU B2 633622 CA AA 2013754 DE C0 69012538 DE T2 69012538 EP A1 436257 EP B1 436257 ES T3 2060004 JP A2 4364103 NZ A 236580 US A 5356628 US A 5368856 US A 5411736	15-09-1994 04-07-1991 04-02-1993 26-06-1991 20-10-1994 12-01-1995 10-07-1991 14-09-1994 16-11-1994 16-12-1992 28-04-1992 18-10-1994 29-11-1994 02-05-1995
EP A1 405291	02-01-1991	AU A1 57759/90 AU B2 619720 CA AA 2016964 DE C0 69011988 DE T2 69011988 DK T3 405291 EP B1 405291 ES T3 2058686 JP A2 3038506 JP B2 2841746 NZ A 234226 PT A 94495 PT B 94495 US A 5001146 US A 5124349 US B1 5124349	03-01-1991 30-01-1992 26-12-1990 06-10-1994 12-01-1995 03-10-1994 31-08-1994 01-11-1994 19-02-1991 24-12-1998 25-06-1992 08-02-1991 28-02-1997 19-03-1991 23-06-1992 20-10-1998
JP A2 6016515	25-01-1994	keine - none - rien	
DE A1 19532447	06-03-1997	keine - none - rien	